10.0 Close

The overall purpose of the Close function is to close all RPP facilities and infrastructure and establish long-term monitoring capability for the Site and the facilities. Closure of tanks and tank farms assumes that waste retrieval will remove sufficient waste from the tanks that the residual waste following retrieval, the tanks themselves, the tank farm ancillary equipment, and the contaminated soil will be disposed of in place according to applicable regulations and agreements. This strategy also assumes that the residual waste and other tank farm source terms will be considered by the U.S. Nuclear Regulatory Commission to be incidental waste, i.e., non-high-level waste. Figure 10.1 shows the portion of the RPP function logic for Closure (ORP 2001c). This function has substantial involvement with studies directed at understanding contaminant migration in the vadose zone and groundwater that are part of the Hanford GW/VZ Integration Project (DOE 1998a, 2000b).

The WBS for the Closure function, expanded to the fourth subfunction/program level, is shown in Figure 10.2. The flowsheet indicating how these subfunctions are conducted is presented in Figure 10.3.

10.1 Key Decisions/Performance Targets for Close

Seven key decisions have been identified for the Close function. The first two key decisions, have been identified high-level key decisions for ORP, as discussed in Section 3. Figure 10.4 presents the SST life-cycle logic to assist in putting these decisions into context.

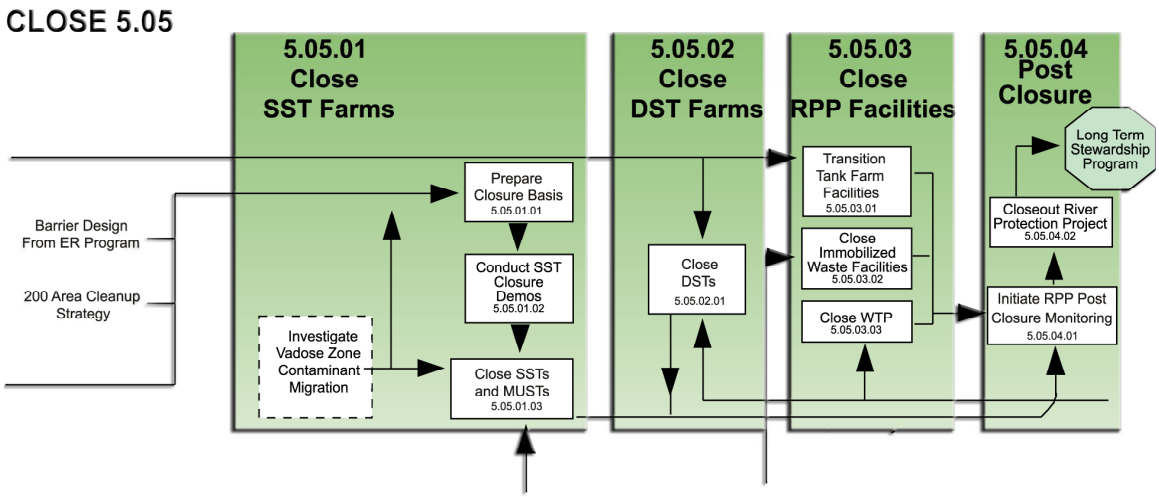

Figure 10.1. Close Functional Logic for the RPP

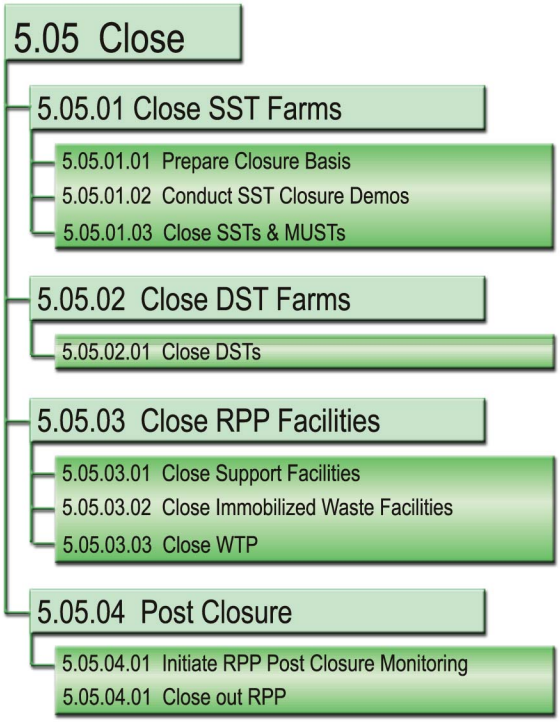

Figure 10.2. Close WBS for the RPP

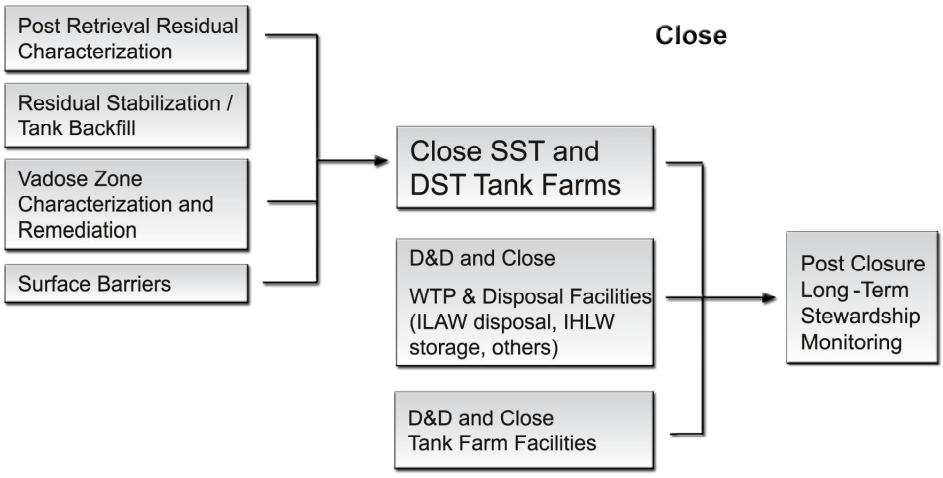

Figure 10.3. Close Functional Flowsheet

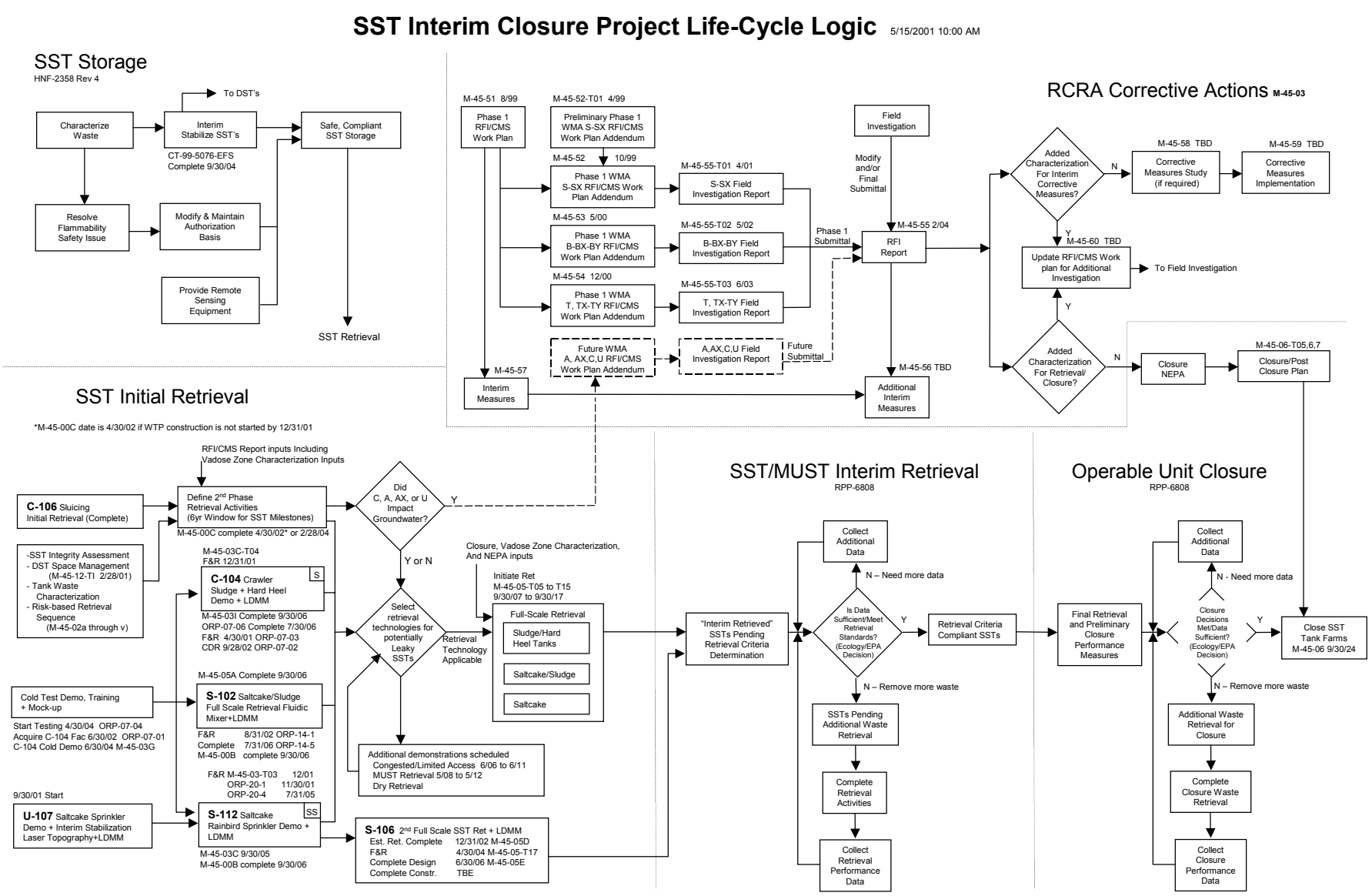

Figure 10.4. Closure Project Life-Cycle Logic

- **Determine Tank Farm Closure Criteria (high-level decision/performance target #14 in Section 3)**. Defining closure criteria for SST farms will require improved understanding of tank residuals and subsurface contaminate transport. This decision is supported by updated closure plans every two years as defined in TPA Milestone M-45-00 (Ecology 1989). The specific technical issue for retrieval is, given retrieval leakage loss, residual tank waste following completion of retrieval, and past tank leak loss, what actions must be implemented to ensure closure of the tank farm in compliance with state and federal regulations. Figure 10.3 describes the SST Interim Closure Project Logic, showing this specific decision. Other key issues center on the release rate and migration of contaminants, including surface and subsurface barriers.

- **Close SST Farms by 2024 (high-level decision/performance target #18 in Section 3)**. The TPA states that the SST tank farms must be closed by 2024. Challenges for closure include measuring post-retrieval inventory, determining how to immobilize residual waste, and selecting and installing surface barriers and long-term monitoring equipment. Furthermore, the negotiation of closure criteria for the SST farms is linked to the outcome and timing of SST retrieval.

- **Determine Whether Data are Sufficient (to show retrieval performance)**. Do data meet retrieval standards for interim retrieved SST? The EPA and Ecology will decide this on a tank-by-tank basis per TPA Milestone M-45-00. See Section 5 for a detailed discussion of tank waste retrieval issues. The specific technical issue is, what, if any, is the maximum residual waste that can remain in a tank following waste retrieval given the 99% interim waste retrieval goals as defined in TPA Milestone M-45-00, technical practicability, compliance with state and federal regulations, and cost. Figure 10.4, which describes the SST interim closure project logic, shows this particular decision under SST/MUST Interim Retrieval.

- **Determine Whether Additional Characterization Is Needed for Interim Corrective Measures**. Figure 10.4 shows this particular decision under RCRA Corrective Actions.

- **Determine Whether Added Characterization is Needed for Retrieval/Closure**. Figure 10.4 shows this particular decision under RCRA Corrective Actions.

- **Determine Whether C, A, or AX Tank Farms Impacted Groundwater**. The leaks from the U tank farms have impacted groundwater. ORP is discussing with Ecology expanding the characterization efforts into all of the SST farms.

- **Determine Approach and Technologies to Complete Tank Farm Closure Based on Established Criteria**. This includes the following components:
  - remediation approach for residual waste in SSTs
  - disposal approach for SSTs
  - contaminated soil remediation approach
  - remediation approach for ancillary equipment
  - surface barriers: performance, settlement and subsidence considerations, and a graded approach to barriers
  - post-retrieval tank waste characterization.

10.2 Risks and S&T Needs Associated with Close

The complete lists of risks identified by the RPP's Risk Management Program as critical risks and critical contractor-internal risks are contained in Appendix B. Consistent with other portions of the process used for this ITP, the risk lists were examined to identify the items associated with closing the tank farms. However, because of the comparatively long-term timeframe for closure decisions, critical risk items from RPP's Risk Management Program do not yet address risks germane to closure of the tanks, tank farms, and infrastructure.

There are several major unresolved issues related to applying risk analysis and risk management in developing and implementing the strategy for closure of the tank farms. Human health risk from remediation and closure activities (short-term risk), and post-closure human health risk and ecological risk (long-term risk) are part of the decision-making process for tank farm closure. In many cases, technology development can provide tools for gathering the information, analyzing the information, or helping provide alternatives with lowered risk.

A recent report from the National Research Council (NRC 2001) said that Hanford must concentrate more on finding cost-effective ways to study, track, and map subterranean contaminants, creating barriers to block those substances from seeping to the Columbia River and prioritizing problems to be tackled.

Those risks that the draft *Single-Shell Tank Closure Interim Project Program Plan*[(a)] identified as being addressed, at least partially, by further S&T development include

- Approach for Evaluating Retrieval Cost/Benefit Tradeoffs. The mechanism has been established and is now being pursued for evaluating cost versus benefit of alternatives for retrieval of waste from SSTs to meet closure requirements, and for selecting the preferred alternatives that will be further developed and deployed.
- Waste Retrieval Required for Closure. The degree of waste removal required for closure should be used as the basis for waste retrieval system technology development, retrieval systems engineering, and definition of completion of retrieval operations. Technology options being pursued involve the following:
  - Salt cake dissolution
  - Movable vehicle-based sluicing
  - Fluidic mixing.
- Technical and Regulatory Basis for Leak Detection Monitoring and Mitigation to Support SST Waste Retrieval and Disposal. A basis (technical and regulatory) needs to be established for the functions and requirements for leak detection and monitoring to support SST retrieval and remediation actions. Current S&T efforts for addressing this include
  - Waste retrieval technology development (including non-hydraulic retrieval system evaluation as the retrieval program progresses)
  - Risk assessments
  - LDMM strategy and technology development
  - Vadose zone characterization.

(a) DOE. 2001. *Draft Single-Shell Tank Closure Work Plan.* DOE-RL-89-16, prepared by CHG for the Richland Operations Office, Richland, Washington.

- Dealing with radioactive and hazardous equipment and materials disposed of in tanks to either permit retrieval of sludge and saltcake or close tanks in accordance with regulatory requirements.
- Potential for differential settlement of the surface barrier across a tank farm.
- Potential for surface barrier subsidence to occur over tanks. Without some form of permanent internal support, tank domes (especially of the larger-diameter tanks) may collapse, leading to rupture of the surface barrier during or after closure because of the weight of existing cover soil and surface barrier materials.

The FY 2002 Hanford RPP Science and Technology Needs http://www.hanford.gov/boards/stcg/needs/tanks.html have been written to document technical aspects of significant risks and uncertainties. The complete list of the RPP's FY 2002 S&T needs is contained in Appendix C. These S&T needs were developed by considering the overall objectives for the Close function and understanding the risks and technical vulnerabilities that have been identified thus far in the programs.

Table I0.I shows the subfunctional structure for the Close function, associated key decisions/performance targets, and how these relate to identified S&T needs and project risks identified to date. This provides a structure for organizing and understanding the interrelationships among these different views of the close function.

10.3 Summary of Technical Activities for Close

Table I0.2 shows how the risks and S&T needs identified for the Close function are related to currently planned technical activities and summarizes schedule and budget information for the technical activities are included. As the table shows, in some cases identified S&T needs have no activities associated with them, and in other cases ongoing technical activities are not associated with specifically identified S&T needs. The S&T needs and the associated technical activities are also summarized in the remainder of this section.

10.3.1 S&T Need RL-WT001, Technetium-99 Analysis in Hanford Tank Waste and Contaminated Tank Farm Area

S&T Need Summary: An accurate production laboratory method for establishing the ^{99}Tc concentration in LLW and vadose zone soils is needed. ^{99}Tc concentration is a critical component of feed to the waste vitrification vendors. The absolute accuracy of these analytical results produced at the Hanford Site has been questioned and found to disagree with results produced at another DOE site. This original issue appears to be resolved based on work in FY I998 for the high-organic-containing waste in which these differences were observed. Variability of redox potential and interferences present in Hanford tank waste can produce inconsistent performance of radiochemical sample preparation methods in use. In addition, the method must be applicable to soils that may contain waste material that leaked from the tank.

Table 10.1. Close Function, Key Decisions/Performance Targets, and Relationship to Risks

Subfunction	Key Decision/ Performance Target	Identified Risks	S&T Need
Close SST Farms 5.05.01 and Close DST Farms 5.05.02	Determine Tank Farm Closure Criteria	Approach for Evaluating Retrieval Cost/benefit Tradeoffs	RL-WT001 - Technetium-99 Analysis in Hanford Tank Waste and Contaminated Tank Farm Area
	Close SST Farms by 2024	Waste Retrieval Required for Closure	RL-WT068 Radionuclide Source Term from Tank Residuals
	Determine Whether Data are Sufficient (to show retrieval performance)	Technical and Regulatory Basis for Leak Detection, Monitoring and Mitigation to Support SST Waste Retrieval and Disposal	RL-WT069 - Value of Information Decision Analysis for Tank Farm Closure
			RL-WT046-S - Getter Materials
	Determine Whether Additional Characterization is Needed for Interim Corrective Measures	Dealing with Radioactive and Hazardous Equipment and Materials Disposed of in Tanks	RL-WT061 - Reactive Barriers to Contaminant Migration
	Determine Whether Added Characterization is Needed for Retrieval/closure	Potential for Differential Settlement of the Surface Barrier Across a Tank Farm	
	Determine Whether C, A, or AX Tank Farms Impact Groundwater	Technical and Regulatory Basis for Leak Detection Monitoring and Mitigation to Support SST Waste Retrieval and Disposal	RL-WT044-S - Distribution of Recharge Rates
			RL-WT035-S - Moisture Flow and Contaminant Transport in Arid Conditions
			RL-WT053-S - Contaminant Mobility Beneath Tank Farms
			RL-WT102 - Advanced Characterization Tools for Contaminants of Concern
	Determine Approach and Technologies to Complete Tank Farm Closure Based on Established Criteria	Potential for Surface Barrier Subsidence to Occur over Tanks	RL-WT017 - Long-life Isolation Surface Barrier
Close RPP Facilities 5.05.03	Close SST Farms by 2024	Waste Retrieval Required for Closure	
Post Closure 5.05.04			

Technetium in the +7 oxidation state is known to be mobile in the soil column; therefore, the concentration in tank waste must be known well to estimate long-term effects of waste tank leakage during storage or retrieval operations. Using inductively coupled plasma/mass spectrometry in place of radiochemical methods may also help resolve some of these chemical issues; however, insufficient data are available to fully support the inductively coupled plasma/mass spectrometry results. Because the inductively coupled plasma/mass spectrometry does not require chemical separations before analysis, it is less subject to the interferences described above. However, other errors may be associated with sample dissolution or polyatomic interferences that have not been clearly defined for this relatively new technology. Finally, when the technology generates data that are used to determine an effect on public/worker safety and health and are critically reviewed by stakeholders, interlaboratory comparisons of the measurement system are needed to raise the level of confidence in the data and credibility of the technology independent of the site at which it is used.

Table 10.2. Technology Activities and Relationships to Identified Risks and S&T Needs for the Close Function (in thousands of dollars)

Key Decisions/ Performance Targets	Identified Risks	S&T Needs	Activities		FY 2001	FY 2002	FY 2003	FY 2004	FY 2005	FY 2006	FY 2007	FY 2008	FY 2009	FY 2010
Determine Tank Farm Closure Criteria Close SST Farms by 2024 Determine Whether Data are Sufficient (to show retrieval performance) Determine Whether Additional Characterization is Needed for Interim Corrective Measures Determine Whether Added Characterization is Needed for Retrieval/Closure	Approach for Evaluating Retrieval Cost/Benefit Tradeoffs Waste Retrieval Required for Closure Technical and Regulatory Basis for Leak Detection, Monitoring and Mitigation to Support SST Waste Retrieval and Disposal Dealing with Radioactive and Hazardous Equipment and Materials Disposed of in Tanks Potential for Differential Settlement of the Surface Barrier Across a Tank Farm	RL-WT001 Technetium-99 Analysis in Hanford Tank Waste	ORP: None TFA: None EMSP: None											
		RL-WT068 Radionuclide Source Term from Tank Residuals	ORP: CHG WBS 5.02.01.01.01.01 SST Retrieval Project Definition TFA: None EMSP: Project 81893 Physiochemical Processes Controlling the Source Term from Tank Residuals Project 81921 Technetium Chemistry in HLW: Role of Organic Complexants Project 81988 Identification of Non-Pertechnetate Species in Hanford Tank Waste, Their Synthesis, Characterization, and Fundamental Chemistry		Δ Report on testing of methods to remove technetium from simulants and real tank heels and solids Δ Report assessing the current knowledge of technetium in alkaline waste heels Δ Report on new experimental testing to determine species and properties of technetium in tank heels and related sludge (at Savannah River) Δ Report on practical limits for technetium removal (identified in B9S1) September, 2002 Δ Complete test of three getter materials with simulant wastes September 2003 Δ Complete tests of promising getter materials with hot wastes funding									
				ORP										
				EMSP										
				81893	$170	$170	$240	$220						
				81921	$115	$295	$230	$310						
				81988	$160	$175	$285	$250						
		RL-WT069 Value of Information Decision Analysis for Tank Farm Closure Activities	ORP: CHG WBS 5.02.01.01.02.02 LDMM (Leak Detection Monitoring & Mitigation) Technology Development TFA: None EMSP: None	ORP	$442	$250	$250							

Table 10.2. (contd)

Key Decisions/ Performance Targets	Identified Risks	S&T Needs	Activities		FY 2001	FY 2002	FY 2003	FY 2004	FY 2005	FY 2006	FY 2007	FY 2008	FY 2009	FY 2010
Determine Whether C, A, AX, or U Tank Farms Impact Groundwater	Technical and Regulatory Basis for Leak Detection Monitoring and Mitigation to Support SST Waste Retrieval and Disposal	RL-WT046-S Getter Materials	ORP: CHG WBS 5.02.01.01.02.02 Retrieval Technology Development	ORP	$200	$200	$200							
			5.04.01.03 Receive and Dispose ILAW											
			5.05.01.01 Prepare Closure Basis				Δ September 2002 Evaluate various retrieval and LDMM activities and make recommendations							
			TFA: A contract has been placed with Sandia National Lab for work exploring the use of an Apatite Reactive Zone. See Section 5.3.5 (RL-WT027 Tank Leak Mitigation Systems)				Δ September 2002 Evaluate minimum level of tank waste cleanup required to meet performance thresholds							
			EMSP: None				Δ September 2003 (as funding permits) Evaluate retrieval and LDMM technologies							
		RL-WT061 Reactive Barriers to Contaminant Migration	ORP: See discussion in Section 10.3.4. (RL-WT046-S)											
			TFA: See discussion in Section 10.3.4.											
			EMSP: See discussion in Section 10.3.4.											
		RL-WT044-S Distribution of Recharge Rates	ORP: CHG WBS 5.04.01.01.04 ILAW Disposal Facility	ORP	$350	$350	$350							
				EMSP										
			TFA: None	70081	$276	$167								
			EMSP: Project 70081, Immobilization of Radionuclides in the Hanford Vadose Zone by Incorporation in Solid Phases	73758	$270	$280	$280							
				70135	$420									
				70219	$417	$343								
			Project 73758, Fixations Mechanisms and Desorption Rates of Sorbed Cs in High Level Waste Contaminated Subsurface Sediments: Implications to Future Behavior and In-Ground Stability	70220	Funding included in Table 5.2									
				70115	Funding included in Table 5.2									
			Project 70135, Colloid-Facilitated Transport of Radionuclides Through the Vadose Zone											

Table 10.2. (contd)

Key Decisions/ Performance Targets	Identified Risks	S&T Needs	Activities		FY 2001	FY 2002	FY 2003	FY 2004	FY 2005	FY 2006	FY 2007	FY 2008	FY 2009	FY 2010
			Project 70219, Fate and Transport of Radionuclides Beneath the Hanford Tank-Fars: Unraveling Coupled Geochemical and Hydrological Processes in the Vadose Zone Project 70220, High Frequency Electromagnetic Impedance Imaging for Vadose Zone and Groundwater Characterization Project 70115, The Use of Radar Methods to Determine Moisture Content in the Vadose Zone											
		RL-WT035-S Moisture Flow and Contaminant Transport in Arid Conditions	ORP: See Section 10.3.8, RL-WT053-S TFA: None EMSP: See discussion in Section 10.3.8.											
		RL-WT053-S Contaminant Mobility Beneath Tank Farms	ORP: CHG WBS 5.05.01.02 SST Closure Demos TFA: None EMSP: Project 81908 Origins of Deviations from Transition-State Theory: Formulating a New Kinetic Rate Law for Dissolution of Silicates See discussion in Section 10.6.3		Δ Sept 01 S&T support for S/X FIR - completed									
						Δ Jan 02 S/X FIR released - completed								
						Δ Oct 02 B/BX/BY FIR released								
							Δ June 03 T/TX/TY FIR released							
				ORP	$1000	$1000	$1000							
				EMSP	$215	$150	$255	$190						
		RL-WT102 Advanced Characterization Tools for Contaminants of Concern	ORP: CHG WBS 5.05.05.01.05 RCRA Corrective Action TFA: None EMSP: See discussion in Section 10.3.6, RL-WT044-S	ORP	$30									
Determine Approach and Technologies to Complete Tank Farm Closure Based on Established Criteria	Potential for Surface Barrier Subsidence to Occur Over Tanks	RL-WT017 Long-Life Isolation Surface Barrier	ORP: CHG WBS 5.02.01.01.05 RCRA Correction Action TFA: None EMSP: None	ORP		$200	$700							
Close SST Farms by 2024	Waste Retrieval Required for Closure		ORP CHG WBS: 5.05.01.02 SST Closure Demos	Funding included in support for RL-WT053-S										
Post Closure 5.05.04														

Failure to develop accurate and reliable measurement methods that are recognized by the regulators, stakeholders, and process operations as confident measurement systems will affect final waste processing requirements and site cleanup criteria. Accurate ^{99}Tc measurements are needed to produce effective risk assessments and ensure that vitrification processes and products will meet acceptance criteria. Failure to meet those criteria could result in additional program costs or regulatory requirements.

ORP Activity: None

TFA Activity: None

EMSP Activity: None

10.3.2 S&T Need RL-WT068, Radionuclide Source Term from Tank Residuals

S&T Need Summary: The Hanford Site composite analysis has shown that the radionuclide source term from residual solids in Hanford HLW tanks is one of the most significant long-term dose contributors on site, especially from ^{99}Tc, ^{79}Se, and uranium isotopes. However, the radionuclide release rate from these solids is virtually unknown. If the release of these important elements can be shown to remain low for time periods of regulatory concern (typically 1,000 to 10,000 years), a higher percentage of residues potentially could be left in the tanks without significant long-term impacts to groundwater and future populations.

Recent chemical analyses of sludges obtained from tanks at Hanford and at SRS suggest that significant fractions of Tc and Se occur in highly insoluble, non-mobile reduced forms in some sludges. Release of Tc and Se from these sludges will be governed by the rate of oxidation of any reduced phase, releasing soluble and highly mobile pertechnetate and selenate anions. Transport of oxidants in the sludge is the likely rate-controlling process. Sludge characteristics/properties affecting permeability to oxidants need to be determined. The effects of proposed tank fillers on sludge aging (phase transformation) and of aging on sludge permeability also need to be determined. A better understanding of the presence and effect of complexants on contaminant mobility is also required.

ORP Activity: CHG WBS 5.02.01.01.01.01. SST Retrieval Project Definition. This activity involves participation in a technical workshop and the preliminary assessment of information on technetium species in the tank heels, supplying samples for TFA testing, and, in the later stages, supplying facilities as well as materials to assist in the testing.

TFA Activity: None

Milestones:

FY01 Report on testing of methods to remove technetium from simulants and real tank heels and solids

FY02 Report assessing the current knowledge of technetium in alkaline waste heels
Report on new experimental testing to determine species and properties of technetium
Report on practical limits for technetium removal (identified in B9S1) September 2002

FY03 Complete test of three getter materials with simulant wastes September 2003
Complete tests of promising getter materials with hot wastes funding

EMSP Activity: Project 81893, Physiochemical Processes Controlling the Source Term for Tank Residuals. Radionuclide release from residual solids in HLW tanks represents a potentially significant source of contaminants migrating in the underlying sediments. A recent composite analysis for the Hanford Site has shown the radionuclide source term from the residual solids is one of the most significant long-term dose contributors on site. However, this assessment was based on a highly conservative release model for the tank residuals. The conservative model is being used in lieu of a true scientific understanding of the processes controlling the release rate from the sludge that is applicable to the Hanford vadose zone environment. The project will significantly improve the fundamental scientific basis for estimating the release rate of ^{99}Tc, the principal long-term dose contributor from tank residual wastes and will develop an improved conceptual model that considers diffusion of water and oxygen in the sludge under conditions of partial hydraulic saturation, but 100% relative humidity, consistent with the subsurface environment at Hanford. Key chemical processes will be considered, including the oxidation of reduced Tc compounds in the sludge and the chemical changes in sludge phase assemblage that will occur over time, in a combination with novel experimental methods to investigate these processes. This comprehensive study will provide a sound technical basis for the DOE and local stakeholders to make more informed cost/benefit/risk decisions regarding closure of Hanford HLW tanks. (FY2001 – FY 2004)

EMSP Activity: Project 81921, Technetium Chemistry in HLW: Role of Organic Complexants. The premise of this project is that Tc complexation with organic compounds in tank waste plays a significant role in the redox chemistry of Tc and the partitioning of Tc between the supernatant and sludge components in waste tanks. These processes need to be understood to develop strategies to effectively remove Tc from high-level nuclear waste prior to waste immobilization and to evaluate long-term consequences of Tc remaining in residual waste after sludge removal. Unfortunately, only limited data are available on the stability of Tc-organic complexes and even less thermodynamic data on which to develop predictive models of Tc chemical behavior. To meet these challenges, this research program will study Tc-speciation in actual tank waste using state-of-the-art analytical organic chemistry, separations, and speciation techniques. On the basis of such studies, thermodynamic data for the identified Tc-organic complexes will be acquired over a wide range of chemical conditions to develop credible models to predict Tc speciation in tank waste and Tc behavior during waste pretreatment processing and in waste tank residuals. (FY 2001 – FY 2004)

EMSP Activity: Project 81988, Identification of Non-Pertechnetate Species in Hanford Tank Waste, Their Synthesis, Characterization, and Fundamental Chemistry. Technetium, as pertechnetate (TcO4-), is a mobile species in the environment. This characteristic, along with its long half-life makes technetium a major contributor to the long-term hazard associated with LLW disposal. Thus, Tc partitioning from nuclear waste tanks at DOE sites may be required so that the LLW forms meet DOE performance assessment criteria. Tc separations assume that Tc exists as pertechnetate in the tank waste. However, work with actual Hanford waste indicates that much of the Tc exists in a form other than pertechnetate and that these unidentified Tc species are not readily converted to pertechnetate by oxidation. This project will use the Tc complexes prepared under the original EMSP proposal to develop a capillary electrophoresis mass spectrometry technique that will be used to identify nonpertechnetate species in actual waste samples. The project has three major goals: 1) develop capillary electrophoresis mass spectrometry as a characterization technique, 2) separate a nonpertechnetate fraction from a waste sample and identify the nonpertechnetate species in it by capillary electrophoresis mass spectrometry, and 3) synthesize and characterize

bulk quantities of the identified nonpertechnetate species and study their ligand substitution and redox chemistry. (FY 2001 – FY 2004)

10.3.3 S&T Need RL-WT069, Value of Information Decision Analysis for Tank Farm Closure

S&T Need Summary: The TWRS EIS (DOE 1996) evaluated waste retrieval and disposal alternatives but did not evaluate tank farm closure alternatives because sufficient information was not available. The ROD for the EIS committed to conducting a NEPA process for decisions on tank farm closure when enough additional information was available on waste retrieval performance, closure technology, and vadose zone conditions. The purpose of waste retrieval from SSTs is to prepare tanks for closure. Until decisions are made on closing tank farms, final requirements for SST waste retrieval cannot be specified, including requirements for allowable residual waste in SSTs after retrieval is completed. Unless a default value of zero leakage during retrieval is specified as an interim requirement, final requirements for leakage control during retrieval also are constrained by decisions on how tank farms will be closed because closure decisions include decisions on remediation of soil potentially contaminated by retrieval leaks. TPA milestones for characterizing the vadose zone in SST waste management areas under RCRA assessment have been established.

Phase I of the required investigations are scheduled to be completed in FY 2004, with recommendations for additional subsurface investigations that may be required to support decisions on waste retrieval and closure. Presently, criteria for making closure decisions (e.g., quantitative measure(s) of compliance, points of compliance, period of compliance, and exposure scenarios) have not been established, although criteria are required to be established under the recently adopted TPA milestones as a basis for determining what subsurface data should be collected. The TPA agreement includes milestones for updating the Tank Farm Closure Work Plans every two years to include results of the "limits of technology" retrieval demonstrations as the information becomes available. By the end of FY 2010, agreement on closure criteria is expected to allow pursuit of the initial Tank Farm Closure Demonstration. Also, no criteria have been established to guide the decision scheduled for FY 2004 on whether additional subsurface data may be needed in a second phase of investigations in the SST waste management areas under RCRA assessment. Because subsurface investigations in contaminated tank farm soils are expensive, an approach is needed for determining when enough information has been gathered to support decisions on closure.

ORP Activity: CHG WBS 5.02.01.01.02.02, LDMM Technology Development. A contract has been placed with Sandia National Laboratory to develop a personal computer-based "decision analysis tool" using a probabilistic framework regarding the extent of necessary field activities, site and waste characterization requirements, and monitoring necessary to support safe and compliant waste retrieval, LDMM, and tank closure based on long-term risks to human health and the environment, regulatory requirements, and stakeholder values. In FY 2001, a probabilistic assessment of the S-112 retrieval system will be performed to address uncertainties and sensitivities in an attempt to reduce conservatism in the Retrieval Performance Evaluation calculations for retrieval release criteria and target leak detection rates.

TFA Activity: None

EMSP Activity: None

10.3.4 S&T Need RL-WT046-S, Getter Materials

S&T Need Summary: Fundamental data are needed to improve confidence in the performance assessment (Mann et al. 2001) under realistic conditions. Negatively charged elements and compounds (e.g., TcO_4^-, Se^-) are poorly sorbed on most materials under basic (pH >7) conditions. However, some negatively charged materials (e.g. I^-) do sorb on Hanford soils under basic conditions. Understanding how important contaminants interact with the soil will help develop appropriate materials to retard the transport of those contaminants.

If low-cost getter materials can be developed for waste disposal, requirements on waste forms can be reduced, potentially saving hundreds of millions of dollars in the Hanford Immobilized Waste Disposal Program. The SRS uses FeS to trap technetium, and many disposal sites use concrete to trap uranium.

This science need supports Hanford tanks technology need, RL-WT061, "Reactive Barriers to Contaminant Migration," discussed in Section 10.3.5.

ORP Activity: CHG WBS 5.02.01.01.02.02, Retrieval Technology Development. 5.04.01.03, Receive and Dispose ILAW, 5.05.01.01, Prepare Closure Basis. General coordination of the contract with Sandia National Laboratory is discussed below in the TFA activities.

TFA Activity: A contract has been placed with Sandia National Laboratory to explore the use of an Apatite Reactive Zone. FY 2001 work involved bench-scale testing of apatite in Hanford soils as a proof-of-concept for sequestering technetium and uranium. The plan is to move to a field-scale demonstration in FY 2002. Costs for the field-scale demonstration will range from $750K to $1,000K in FY 2002. Pending a successful field-scale demonstration of the Apatite Reactive Zone concept during FY 2002, an actual deployment may occur in the 200 West Area groundwater, near S-SX Tank Farm, where elevated levels of technetium have been found. (See also discussion associated with Need RL-WT068 in Section 10.3.2 and the TFA Strategic Initiative B9S1, "Controlling Radionuclide Source Terms Important to Tank Closure.") See Section 5.3.5.

Milestones:

Task 1, September 2001: Probabilistic assessment of the S-112 retrieval system.

Task 2, September 2002: Evaluate various retrieval and LDMM activities and make recommendations

Task 3, September 2002: Evaluate minimum level of tank waste cleanup required to meet performance thresholds

Task 4, September 2003: (as funding permits) Evaluate retrieval and LDMM technologies

EMSP Activity: None

10.3.5 S&T Need RL-WT061 Reactive Barriers to Contaminant Migration

S&T Need Summary: Although the SSTs and DSTs store a broad range of highly radioactive isotopes, a few relatively mobile constituents dominate the risk to human health and the environment. For the vadose zone groundwater pathway, based on past analysis, the constituent list typically includes ^{99}Tc, ^{129}I, ^{79}Se and U. The relative importance of these constituents varies depending on assumptions used during the specific analysis.

Of the 149 SSTs at the Hanford Site, 67 are known or suspected leakers. Retrieval of waste from these tanks will incur risk from additional leakage. In addition, waste that has been retrieved will be processed, vitrified, and disposed in solid form. Based on past analyses, this waste may add radionuclides to the soil column. For example, the performance assessment activities supporting the disposal of vitrified LAW ^{99}Tc and ^{79}Se as the radionuclides that contributed most significantly to long-term risk. If these key radioactive elements could be trapped or immobilized in the waste matrix, disposal facility, closed tanks, and/or the soil column, the risk to human health and the environment could be significantly reduced. It has been proposed to deploy sequestering agents as a permeable flow-through (reactive) barrier to attenuate the migration of these contaminants and reduce the risk. In the case of contaminated soil, the reactive barrier will be placed using conventional emplacement technology, e.g., slant drilling. For the vitrified waste and tank closure, it has been proposed that the getter could be placed inside the facility. For existing waste sites, the material may need to be injected into the soil underlying the facility.

Although limited efforts have been performed to identify getter materials (sequestering agents), no material has been sufficiently tested to date to be selected. During the last few years, the list of candidate materials has been reduced. Based on this work, candidate getters include bone char, hydrotalcite, iron oxyhydroxides, sulfides, magnetite, and oxides. Research to date (performed by both PNNL and Sandia National Laboratory) suggests magnetite, bone char, and hydrotalcite to be most effective for attenuating technetium. Similarly, hydrotalcite and iron oxyhydroxides are candidates for attenuating uranium and selenium. Recent efforts as part of the Immobilized Waste Program and by the TFA champion have identified some potentially useful materials.

ORP Activity: See the discussion in Section I0.3.4, "Getter Materials."

TFA Activity: See the discussion in Section I0.3.4, "Getter Materials."

EMSP Activity: See the discussion in Section I0.3.4, "Getter Materials."

10.3.6 S&T Need RL-WT044-S, Distribution of Recharge Rates

S&T Need Summary: There is a need for fundamental data to improve confidence in estimating recharge rates as a function of time and space for use in impact assessments under realistic conditions.
The rate at which moisture exits the surface root zone of soil and enters into the subsurface (often called the recharge rate) is often the major parameter (along with inventory) determining the rate at which contaminants enter groundwater, particularly in dry climates. The recharge rate is known to depend on many parameters (e.g., type and condition of surface soil, type and extent of vegetation, and climate). However, this dependence is usually determined for idealized conditions and for small spatial and temporal extents. For large sites such as Hanford, where waste disposal covers many acres and impact calculations must extent beyond thousands of years,

such simple descriptions are inadequate to convince the technical community, the regulators, and the stakeholders that impacts can be adequately estimated.

Short-term measurements of natural recharge rates at local scales at the Hanford Site range from less than I mm/yr to basically the precipitation rate (I60 mm/yr). Longer-term measurements (e.g., tracer measurements) have shown a greater variability. Man-made effects (e.g., funneling) as well as topographic features can enhance these values. Furthermore, Hanford Site impacts assessments (e.g., *Hanford Immobilized Low-Activity Tank Waste Performance Assessment: 200I Version*) (Mann et al. 200I) have shown the dependence of impacts on recharge rate. Such conclusions are reinforced by Hanford work for the Nuclear Regulatory Commission on the importance of surface barriers.

ORP Activity: CHG WBS 5.04.01.01.04, ILAW Disposal Facility. This activity is supporting PNNL's measurements of recharge under various soil and vegetation conditions at the Field Lysimetry Test Facility, PNNL's model development and validation for Hanford conditions, and PNNL's data collection for model input parameters.

TFA Activity: None

EMSP Activity: EMSP Project 70081, Immobilization of Radionuclides in the Hanford Vadose Zone by Incorporation in Solid Phases. This multi-institutional project will investigate whether soluble radionuclide contaminants in soils, groundwater, and tank wastes sorb to or desorb from particle surfaces. When sorbed as surface complexes, such contaminants are susceptible to changes in solution pH and ionic strength, which can cause contaminants to be desorbed and mobilized. However, incorporating the contaminant in stable insoluble solids can immobilize and isolate it. This process may be useful in treating contaminants leaked from underground storage tanks at Hanford. The high concentrations of dissolved Al and exceptionally high pH (>I3) and ionic strengths of tank fluids make it probable that leachates reacting with the soil will form complex precipitates that could incorporate contaminant species from the tanks. Tank fluids reacting with the soil should dissolve some of the soil minerals, decreasing the pH. This project will perform laboratory studies on precipitation processes occurring in the Hanford vadose zone, characterizing the particle coatings and precipitate phases in core samples. These investigations will use x-ray-absorption fine structure, nuclear magnetic resonance, and vibrational spectroscopies, electron and x-ray microprobe analyses, transmission electron microscopy, X-ray photoelectron spectroscopy, and other characterization methods sensitive to the speciation and spatial distribution of these ions in several model systems. (FY I999 – FY 2002)

EMSP Project 73758, Fixations Mechanisms and Desorption Rates of Sorbed Cs in High Level Waste Contaminated Subsurface Sediments: Implications to Future Behavior and In-Ground Stability. This project will apply new understanding of cesium geochemistry and newly developed single-particle manipulation/analytical capabilities to cesium-contaminated sediments leaked from SSTs at Hanford. Because the leaks occurred 25 to 35 years ago, long-term fixation processes and their implications can be defined. The project will study the mineralogic residence and intraparticle distribution of ^{137}Cs in sediments contaminated with HLW and will define the relationship of residence and distribution to ^{137}Cs desorbability and exchange with the aqueous phase. Contaminated sediments will be studied from boreholes beneath leaked tanks. Microparticle manipulation and analytical techniques (e.g., low-level gamma counting, digital auoradiography, transmission electron microscopy with energy loss spectroscopy, electron microprobe, dynamic and time-of-flight secondary ion mass spectrometry, and synchrotron x-ray microspectroscopy) will be applied to isolate and quantify spatial distribution of Cs in the contaminated particles. Desorption kinetic studies will be performed on model and contaminated sediments with

different chemical conditions and a model developed that links kinetic behavior with Cs-containing particle characteristics. The resulting understandings of the geochemical behavior of sorbed Cs in the vadose zone will help develop remediation plans for the tank farms. (FY 2000 – FY 2003)

EMSP Project 70135, Colloid-Facilitated Transport of Radionuclides Through the Vadose Zone. This project will study the major processes responsible for colloid-facilitated transport. Colloid-facilitated transport is thought to occur at Hanford, where contaminants leaking from the storage tanks are released directly to the vadose zone and the potential for in situ formation of colloids is high. Formation of colloids will be studied by reacting typical tank waste solutions with vadose zone materials and solutions expected at equilibrium with soil material. The interactions of cesium with colloidal particles will be investigated. Transport of colloids and cesium will be studied using Hanford sand. Magnetic resonance imaging will be used to visualize colloidal movement inside the porous medium. Sorption studies and column outflow data will be analyzed with numerical models to elucidate mechanisms responsible for contaminant sorption as well as colloid and radionuclide transport. Sorption and reaction models will be combined with transport models to quantitatively describe the column experiments. The results will lead to a better understanding of colloid-formation, colloid-contaminant-soil interactions, colloid migration, and colloid-facilitated transport in the vadose zone. The results are directly applicable to cleanup strategies for Hanford. (FY 1999 – FY 2002)

EMSP Project 70219, Fate and Transport of Radionuclides Beneath the Hanford Tank Farms: Unraveling Coupled Geochemical and Hydrological Processes in the Vadose Zone. The goal of this research is to understand coupled hydrological and geochemical mechanisms responsible for accelerated migration of radionuclides in the vadose zone beneath the Hanford tank farms. Field-relevant, long-term unsaturated flow and transport experiments in undisturbed Hanford sediments will be performed that involve 1) multiple tracer strategies for quantifying preferential flow and nonequilibrium mass transfer processes at various water contents, and 2) several novel surface analyses techniques (e.g., x-ray computed tomography, x-ray absorption spectroscopy, hyperquenching fluorescence) to quantify the distribution and chemical environment of contaminants as a function of sediment lithology and water content. The results from this research will provide information in previously unexplored areas of vadose zone fate and transport to support DOE-EM's performance/risk assessment and decision-making process for tank farm restoration. This work combines environmental restoration and major user facilities (i.e., Stanford Synchrotron Radiation Laboratory, Advanced Photon Source, Environmental Molecular Sciences Laboratory) as well as academia. (FY 1999 – FY 2002)

EMSP Project 70220, High Frequency Electromagnetic Impedance Imaging for Vadose Zone and Groundwater Characterization. This project is summarized in Section 5.3.4 as part of leak detection during retrieval of SST waste.

EMSP Project 70115, The Use of Radar Methods to Determine Moisture Content in the Vadose Zone. This project is summarized in Section 5.3.4, "Tank Leak Mitigation Systems" as part of leak detection during retrieval of SST waste.

10.3.7 S&T Need RL-WT035-S, Moisture Flow and Contaminant Transport in Arid Conditions

S&T Need Summary: The movement of contaminants through zones of low moisture (region-wide saturation less than 10%) needs to be understood for risk assessments. Most of the work on moisture flow and

contaminant transport has been done at sites important for agriculture, i.e., sites having moisture contents near saturation. Thus, the theories and equations for moisture flow and contaminant transport are modifications of theories and equations for fully saturated environments. In such an environment, movement through the pore spaces between soil particles is dominant. Under very dry conditions, the interactions with the soil particles become more important. Tested theories and equations are needed for performance assessments to predict moisture movement and contaminant transport. Without this information, conservative methods and data will be used in performance assessments, likely requiring more stringent contaminant release specifications in the waste product.

ORP Activity: See discussion in Section I0.3.8, "Contaminant Mobility Beneath Tank Farms."

TFA Activity: None

Milestones: See discussion in Section I0.3.8, "Contaminant Mobility Beneath Tank Farms."

EMSP Activity: See EMSP Projects discussion in Section I0.3.6, "Distribution of Recharge Rates."

10.3.8 S&T Need RL-WT053-S Contaminant Mobility Beneath Tank Farms

S&T Need Summary: The evolution of the present distribution of contaminants, both radioactive and nonradioactive (particularly ^{137}Cs, but also Pu, ^{99}Tc, ^{90}Sr, Cr, and nitrate), beneath the tank farms needs to be quantified and understood and their potential mobility evaluated. The current understanding of the mobility of contaminants from SST leaks and major soil column transuranic disposal sites is inadequate to fully support cleanup, closure, or performance assessment-related decisions. Notably, borehole logging in SX Tank Farm revealed ^{137}Cs at depths of 40 meters (I30 ft), significantly deeper than predicted by current models. Further investigations, including the drilling of two additional wells, confirmed the presence of migrated cesium in the formation. The report issued by the RPP Vadose Zone Expert Panel concluded that cesium migration was poorly understood and that insufficient data were available to validate migration models.

Without knowledge about the distribution of contaminants beneath the tank farms and the ability in hand to predict contaminant movement, the public cannot be assured that DOE can predict the impact of leaks during sluicing of the tanks during cleanup and the impact of leaving the tanks (and their associated subsurface contamination) in place.

Furthermore, the vadose zone cleanup schedule for the 200 Areas could be delayed if the mobility status of deeply distributed contaminants is unknown or inadequately characterized well in advance. For example, if it is eventually determined that retrieval of TRU-contaminated soil down to 40 m or more beneath Plutonium Finishing Plant cribs is required, the cleanup schedule could be greatly impacted by excavation and handling costs that could approach I billion dollars or more. Similar excavation requirements for leaking SSTs could drive the costs of cleanup higher by several orders of magnitude. The sooner this issue is resolved, the sooner more accurate technical, financial, and schedule forecasts can be made

ORP Activity: CHG WBS 5.05.01.02 SST Closure Demonstrations. The Ground Water Vadose Zone Integration Project provided much support to ORP in FY 200I. Three primary tasks provide support to ORP that are funded directly through the Integration Project: I) field investigations at representative

sites, 2) transport modeling, and 3) vadose zone transport field studies. RPP (CHG) staff recognize the strong linkages of the first two tasks with their work because S&T products feed directly into their field investigation reports.

Field Investigations at Representative Sites. Scientific study is being focused on core materials collected as part of tank farm assessment activities. The science activities are to complement, or "wrap around" physical and chemical characterization measurements to be made on these same materials by ORP for determining contaminant distribution and subsurface conditions. The wrap-around science applies state-of-the-art concepts and analytical methodologies to advance understanding of subsurface water and contaminant migration processes (past and future) beneath leaked SSTs. The overall objectives of the wrap-around science are to develop sound conceptual models (e.g., linked processes) of waste migration beneath key site types as needed by the RPP, System Assessment Capability, and other components of the Integration Project (e.g., systems characterization function) and to provide a basis for developing improved contaminant transport models and credible strategies for corrective action and waste site closure.

Transport Modeling. The objective of the transport modeling task is to provide vadose zone contaminant transport model(s) that adequately incorporate new knowledge derived from investigations at representative field sites, waste/sediment laboratory experiments (e.g., performed through EMSP), and vadose zone transport field studies.

Vadose Zone Transport Field Studies. The objective of this task, which will be conducted at uncontaminated sites, will be to collect data sets to verify conceptual and numerical models that describe transport through the vadose zone. Results will be used in the System Assessment Capability, Rev. I, and in the RPP Field Report for the waste management area involving tank farms B-BX-BY. A field investigation strategy and a broad outline of multi-year activities have been prepared for this task. The broad outline provides input for annual updates to the groundwater/vadose zone S&T Roadmap.

TFA Activity: None

EMSP Activity: See also the discussion on EMSP projects in Section I0.3.6, "Distribution of Recharge Rates."

Project 81908, Origins of Deviations from Transition-State Theory: Formulating a New Kinetic Rate Law for Dissolution of Silicates. Contaminant release and migration models that are used to study a broad range of geochemical and environmental science problems commonly employ transition-state theory to regulate the dissolution kinetics of waste forms and minerals. Despite the widespread use of transition-state theory in sophisticated reaction-transport codes, recent experiments have revealed that models based on such arguments are inaccurate when modeling solutions that contain high concentrations of dissolved components. Because solutions in subsurface environments are typically near saturation, transition-state-theory-based predictions are questionable and so may be remediation schemes that stem from these projections. Therefore, overly conservative and costly designs for high- and low-level waste disposal sites and remediation strategies for plume migration will have to be pursued to circumvent errors. This project will develop and validate a new rate law that more accurately predicts dissolution kinetics in realistic solution compositions. The reactivity of borosilicate glass, synthetic plagioclase feldspar, and biotite will be determined over a range of solution saturation values. Because the solutions will be doped in the same elements that comprise the solids, the solid phases will be doped with radioactive tracers. To determine the control of element release in near-saturated solutions on defect populations, atomic force microscopy and vertical scanning

interferometry will be used to compare rates of natural and synthetic feldspar and biotite. Surface species will be quantified using specular reflectance and photoacoustic Fourier Transform Infrared Spectroscopy methods. From these data the reactive precursor species that are the key to unraveling rate mechanisms will be identified. The resulting model will have widespread application to geochemistry problems and will form the foundation of sound prediction. (FY 200I – FY 2004)

10.3.9 S&T Need RL-WT102, Advanced Characterization Tools for Contaminants of Concern

S&T Need Summary: The extent of contamination in soil, burial ground, and tank farm sites is often poorly defined. A cost-effective technology that provides real-time, in situ measurement of radioactive (technetium, uranium, plutonium, strontium-90, iodine, and selenium) and hazardous (chromium, mercury, lead, nitrate, sodium) contaminants of concern in soils at depth is required to better define the contaminant plume boundaries prior to remediation and also to support long-term monitoring for performance validation of the completed remediation activity. A recent report of the NRC (200I) cited this as one of the major S&T needs fore the Hanford Site.

ORP Activity: CHG WBS 5.05.05.01.05 RCRA Corrective Action. This activity includes developing high-efficiency sampling tool for highly radioactive areas. Details are discussed in Section I0.3.2, "Radionuclide Source Term from Tank Residuals."

TFA Activity: None

EMSP Activity: See the EMSP project discussion in Section I0.3.6, "Distribution of Recharge Rates."

10.3.10 S&T Need RL-WT017, Long-Life Isolation Surface Barrier

S&T Need Summary: Surface barriers are being used over many Hanford waste sites contaminated with low-level radionuclides and/or chemical contaminants. Many more waste sites are expected to use such barriers in the future. These barriers are used to reduce moisture infiltration and plant and animal intrusion. Short-term testing of barriers has occurred under project-sponsored activities, but long-term studies remain a funding orphan. Because the design life of the barrier is 500 to 1,000 years, data will be needed on degradation to better understand the validity of the design life estimate. Concern exists regarding the integrity of barrier designs and the definition of adequate testing to verify barrier performance. This technology need relates to the generation and subsequent regulatory acceptance of adequate design, selection, validation, and monitoring results. Acceptance of these results will allow an environmentally sound, cost-effective, graded design approach for barrier implementation at the Hanford Site. The recent report of the NRC (NRC 200I) cited this as the major S&T need for the Hanford Site.

ORP Activity: CHG WBS 5.02.01.01.05 RCRA Corrective Action. This activity involves developing requirements for barriers inside active tank farms and designing and installing install interim barriers (20 to 30 years) in one or more tank farms.

TFA Activity: None

EMSP Activity: None